

REPORT DOCUMENTATION PAGE

Form Approved
OMB NO. 0704-0188

Public Reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comment regarding this burden estimates or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave Blank)		2. REPORT DATE June 30, 2000		3. REPORT TYPE AND DATES COVERED Final Report: 06/95 to 11/99	
4. TITLE AND SUBTITLE Novel Electric-Field Effects in Quantum Wells, Superlattices, and Microcavities				5. FUNDING NUMBERS DAAH 04-95-1-0532	
6. AUTHOR(S) Emilio E. Mendez					
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) State University of New York at Stony Brook Stony Brook, NY 11794-3800				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) U. S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211				10. SPONSORING / MONITORING AGENCY REPORT NUMBER <i>ARO 34918.1-CH</i>	
11. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.					
12 a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution unlimited.				12 b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) We have used an external electric field to control important properties of semiconductor quantum wells. Specifically: 1) We have modified the exciton-exciton interaction. This interaction is determined by many-body effects, which depend on the overlap between the exciton's electron and hole wavefunctions, and is manifested in the energy difference between photoluminescence spectra with different polarizations. In time-resolved photoluminescence (PL) experiments with circular polarization we have observed that spectral difference, which we controlled by an electric field applied to GaAs-GaAlAs coupled quantum wells. Our results confirm the predictions of theory but also point out its limitations to fully explain our observations. 2) We have reversed the valence-band ordering in strained-layer quantum wells, thus "undoing" the effects of strain. To prove the concept we have used strained InGaAs-InAlAs quantum wells in which the light-hole state is the ground state in the valence band. Photocurrent measurements under various fields have shown a change in the valence-band states that contribute to the fundamental (lowest-energy) transition, from light-hole states to heavy-hole states. This result opens the door to reversing the polarization of light emission in quantum wells, from TM to TE, which could find application in optical modulators. 3) We have shown the presence of low-temperature exciton-photon coupling in microcavities using PL spectroscopy. Because of thermalization until now it has been almost impossible to use PL to study at low temperature (T = 20K or less) the coupling of excitons and photons. We have determined the difference between the lowest-energy PL peak from a quantum well-microcavity system and that of an isolated quantum well, at various temperatures. This difference is constant with T when a field is applied but is T dependent when the field is suppressed. The maximum variation is a direct measure of the exciton-photon coupling.					
14. SUBJECT TERMS Semiconductor Heterostructures, III-V Compounds Electric-field Effects on Optical Properties, Excitons in Quantum Wells Quantum Wells, Superlattices, Microcavities Optical Modulators				15. NUMBER OF PAGES 10	
				16. PRICE CODE	
17. SECURITY CLASSIFICATION OR REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION ON THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL		

NSN 7540-01-280-5500

Standard Form 298 (Rev.2-89)
Prescribed by ANSI Std. Z39-18
298-102

20001122 112

REPORT DOCUMENTATION PAGE (SF298)

(Continuation Sheet)

Novel Electric-Field Effects in Quantum Wells, Superlattices, and Microcavities

(Period: August 1995 to November 1999)

Introduction

It is now almost two decades since the first proposal and demonstration of the effects of an external field on the electronic properties of quantum wells. During this time, those effects have been exploited in optoelectronic applications (for instance, in electro-optic modulators) and have been used to elucidate important physical concepts (e.g., the Wannier-Stark ladder in superlattices). In spite of the relative maturity of the subject, new physics ideas keep coming to light and new possibilities for applications keep appearing. In the following, we highlight some of the concepts developed along those lines as part of this ARO grant in the 1995-99 period. Rather than describing in detail all the projects we have been involved with during the tenure of this grant, we focus here on the work that is already completed and that represents the core of the initial proposal. Also initiated during that period is our preliminary work on optical gain in intersubband cascade lasers based on type II heterostructures and on-going work on time-resolved semiconductor microcavities, both of them described in previous annual reports.

Accomplishments

1. Tuning by an Electric Field of the Exciton-Exciton Interaction in Quantum Wells

The two components of a spin-polarized exciton gas have different energy, as a result of a density- and spin-dependent modification of the two dimensional exciton binding energy by two competing processes: the exchange interaction between electrons (and holes) of different excitons, and the vertex correction to the Coulomb interaction between electrons and holes of different excitons, which effectively reduces the electron-hole attraction. The energy splitting between the two components, ΔE , has been observed in the past in time-resolved photoluminescence (PL) experiments in quantum wells, in which a circularly polarized light pulse creates excitons with spin +1 that gradually relax into the spin -1 state, until both populations are equal.

However, until now no experiment had tested the theoretical prediction of a decrease or even a sign reversal in ΔE caused by a modification of the vertex correction and the exchange term

when the overlap of electron and hole wavefunctions is reduced. We have obtained the first experimental evidence of such a change, determined from measurements of ΔE in coupled double quantum wells (CDQW) subjected to a longitudinal electric field that separates spatially the electrons and holes. We not only find a good agreement between the observed energy splitting and the predicted one, but our results go beyond available models. Specifically, we have found that screening of these many-body corrections is strongly reduced by the field, an effect not contemplated by theory so far.

The material structure used for the experiments was an $n^+ - i - p^+$ GaAs-AlGaAs junction. The active region consisted of ten CDQWs periods separated from each other by a 200\AA $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layer, and each period formed by two 50\AA GaAs wells with a 20\AA $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barrier in between. An undoped $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layer on each side of the CDQW stack (1000\AA and 80\AA thick, respectively) completed the intrinsic (i) region of the structure, whose p^+ and n^+ electrodes were a 5000\AA GaAs:Be layer and a $1\text{ }\mu\text{m}$ thick GaAs:Si layer on a $[100]$ GaAs substrate, respectively.

Time-resolved PL measurements (at $T = 8\text{K}$) were performed at Universidad Autonoma de Madrid (where the Principal Investigator visited during June-July of 1998 and January of 1999) using a standard up-conversion set-up that included a Ti:Sapphire laser with 2 ps pulse width. For polarization-selective excitation and PL detection, $\lambda/4$ wave plates were placed in the paths of the laser beam and the PL collecting optics.

Our focus has been on the two spin components of ground-state electron-heavy hole excitons $X\{\text{e1hh1}\}$. Initially a $+1$ exciton population was created; however, spin-flipping processes gave rise to a non-equilibrium mixture of $+1$ and -1 excitons with populations N_{X+} (majority) and N_{X-} (minority), respectively. The degree of polarization, defined as $P = \{N_{X+} - N_{X-}\} / \{N_{X+} + N_{X-}\}$, was found to be initially $P_0 = 0.50 \pm 0.05$, independent of the electric field. A measure of the exciton-exciton interaction is the energy difference ΔE between the PL spectra of the X^+ and X^- excitons.

The electric-field dependence of ΔE is summarized in Fig. 1. The maximum splitting of 4 meV is reached at zero field, and then it decreases linearly until it becomes zero at a field of $E = 35\text{ kV/cm}$. The parameter d in the upper scale of Fig. 1 represents the average separation (along the growth direction) between electron and hole as calculated from the field dependent expectation values. The zero splitting is reached at an e-h separation of 60\AA , which is a factor of 2.5 larger than the value predicted by the only theoretical model available. This discrepancy is not surprising in view of the simplifications of that model, such as zero temperature and a strictly-2D confinement.

An analysis of the power dependence of the ± 1 exciton's energies at a short delay time, and hence at constant polarization, allows a direct comparison of the experiments with theory and a test of its underlying assumptions. At low fields ΔE increases considerably with power, in agreement with previous experimental results at zero field and with the predictions of theory. At

intermediate fields $E = 23$ kV/cm, the splitting still grows with increasing power, however the values of ΔE are reduced with respect to the low field case, as a consequence of the enhanced e-h separation. At a field $E = 35$ kV/cm, ΔE remains zero at all excitation powers, in agreement with the theoretical model, which predicts that at a certain d the vertex and exchange corrections become equal, independently of the total exciton population.

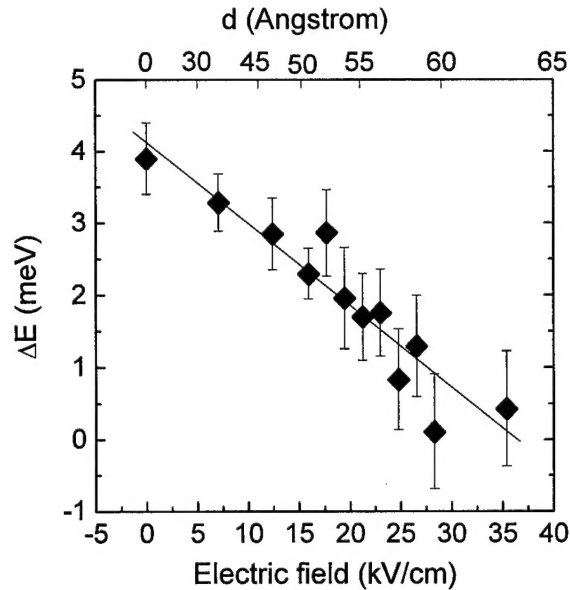


Fig. 1. Energy splitting between the + and - exciton components as a function of electric field. The upper axis represents the electron-hole separation along the growth direction. The solid line is a linear fit to the data.

2. Valence Band Order Controlled by Electric Field in Strained Quantum Wells

Optical modulators based on quantum wells and superlattices have very desirable properties for two-dimensional photonic switch arrays used in high speed fiber interconnects and optical parallel processing: they are fast, planar, and compatible for optical integration with devices such as lasers, detectors, and transistors. A possible modulation scheme consists on switching the polarization mode of the electromagnetic radiation, either from the TE to the TM mode or viceversa. Since the predominant optical transition in semiconductor quantum wells usually involves a heavy-hole state from the valence band, most quantum optical devices are transverse electric (TE) polarization functional.

To build a TM wave dominant structure, the light hole level must overcome the splitting created by the quantum-size effect which favors heavy hole. This condition can be achieved by applying tensile strain to the quantum well. Compressive strain decreases the heavy hole bandgap and increases the light hole bandgap whereas tensile strain produces the opposite effect. Thus, under tensile strain the separation between the heavy- and light-hole subbands is reduced,

and given enough strain the light-hole subband may become the lowest band, so that optical emission or absorption will have TM character.

When an electric field is applied to a quantum well, the electron-hole transition energy exhibits a red shift and the corresponding oscillator strength decreases because the electron and hole wavefunctions are separated by the field. The amount of shift depends on the effective mass: the larger the mass, the larger the shift. Therefore, in a quantum well in which the light hole is the lowest energy level relative to the top of the valence band, the heavy hole can become the lowest level when a sufficiently large electric field is applied. A field-induced TM-to-TE polarization switch becomes then possible: at zero or low external bias the optical absorption of the quantum well is TM active, then at high bias it is transformed into TE active.

To study the viability of these ideas, we chose InGaAs/InAlAs heterojunctions grown on InP substrates. A quantum well composed of an $\text{In}_x\text{Ga}_{1-x}\text{As}$ region and $\text{In}_y\text{Al}_{1-y}\text{As}$ barriers with $x < 0.53$ and $y > 0.52$ is able to move the light hole band close to the conduction band and heavy hole band away from the conduction band because the lattice constant of $\text{In}_y\text{Al}_{1-y}\text{As}$ is larger than that of $\text{In}_x\text{Ga}_{1-x}\text{As}$. In our study, we have designed three structures in which the wells are subjected to either no strain (sample A: 150\AA $\text{In}_{.53}\text{Ga}_{.47}\text{As}$ well with 100\AA $\text{In}_{.52}\text{Al}_{.48}\text{As}$ barriers), moderate tension (sample B: $\text{In}_{.50}\text{Ga}_{.50}\text{As}/\text{In}_{.57}\text{Al}_{.43}\text{As}$), and large tension (sample C: $\text{In}_{.48}\text{Ga}_{.52}\text{As}/\text{In}_{.60}\text{Al}_{.40}\text{As}$) to reverse the valence band order. The samples, capped with p-type regions were grown by molecular beam epitaxy on n-type (100) InP substrates at the Army Research Laboratory (Richard Leavitt's group).

Figure 2 summarizes the lower transition energies for samples A, B, and C obtained from the excitonic peaks which are apparent in photocurrent measurements at $T = 100\text{K}$. The identification of the various peaks was possible only after careful numerical calculations were performed to determine the various excitonic interband transitions in the presence of an electric field. The envelope function approximation was used to solve the Schrödinger equation and the finite-difference method was chosen in order to incorporate the electric field easily in the calculation. When calculating the heavy hole and light hole energy levels, the effect of strain was included by adjusting the valence band offset according to the corresponding strain.

The response of the three samples looks alike except for some differences on the low-energy side. For sample A, the peaks at 830 meV and 840 meV when $V = -1\text{V}$ originate in the E1HH1 and E1LH1 transitions respectively, as it corresponds to a non-inverted band alignment. On the other hand, for the same voltage the lowest transitions in sample C are at 810 meV and 820 meV, and this time they correspond to the E1LH1 and E1HH1 transitions respectively. In this case, in which the bands are inverted at zero or low field, we observe that when the bias increases the two lines cross each other at about 3V. Then, at even higher fields, the E1LH1 transition loses strength and is not visible beyond -7V .

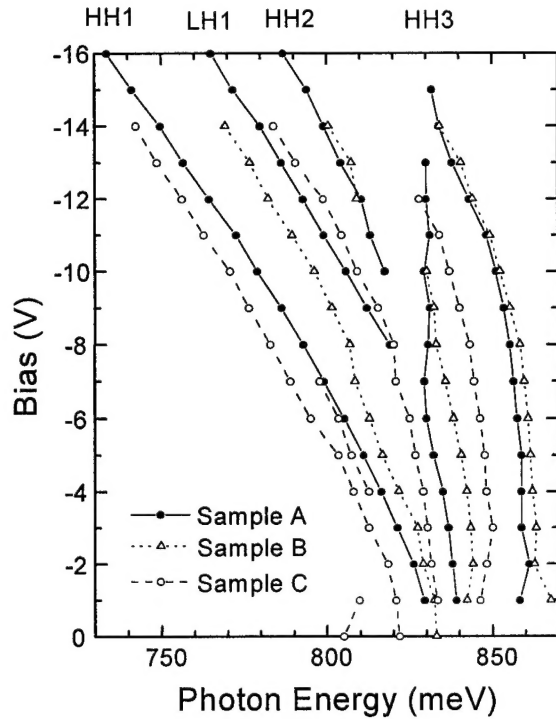


Fig. 2. Dependence of the peak energies of photocurrent spectra (taken at $T = 100\text{K}$) on the external voltage applied to three samples consisting of InGaAs/InAlAs quantum wells of slightly different composition.

In sample B, with a tensile strain intermediate with those of A and C, the E1HH1 and E1LH1 transitions are essentially at the same energy for $V = -1\text{V}$ at an energy between those of E1HH1 and E1LH1 for sample A. Based on the electric field dependence, at low fields the lowest-energy transition has a predominant E1HH1 character. However, in the high-field regime ($V > -8\text{V}$), it is the E1L1 transition that dominates. This peculiar behavior may be a consequence of the strong mixing between the neighboring heavy and light hole bands, although it does not seem to explain the high-field regime, where that coupling should be reduced.

Our results demonstrate that by combining tensile strain and Stark effects a polarization controllable optoelectronic device should be feasible.

3. Electric Field Control of Exciton-Photon Coupling in Semiconductor Superlattices

In a semiconductor microcavity with an imbedded quantum well, when the characteristic energy of the cavity mode coincides with that of the quantum well exciton (resonance condition) there is a strong coupling between the two, provided the exciton's oscillator strength is sufficiently large. In analogy to excitons in bulk semiconductors, the dispersion relation of the coupled exciton-photon system, termed polariton, has two energy branches. These branches have a mixed-mode character, with the relative exciton/photon mix of each branch being dependent on the amount of coupling. Since the individual decay times of a cavity mode and an exciton are very different (about 1 ps the former and several hundreds of ps the latter), the overall light emission lifetime depends strongly on the degree of coupling. Moreover, the relative distribution of a polariton population (created, e.g., by optical absorption) between both branches will depend on the decay times of each one and on the momentum relaxation of the polaritons to the bottom of each branch.

Because in semiconductors the emission wavelength λ varies with temperature whereas the layer thickness does not, resonance between the well exciton and the photon mode of the cavity is achieved only for a very small temperature range. We have used this fact to demonstrate with photoluminescence spectroscopy the existence of strong exciton-photon coupling at moderately high temperatures ($T = 120\text{K}$). In a broad range of temperatures around a characteristic temperature T_0 , the PL spectrum consists of two peaks corresponding to the exciton and cavity energies and whose separation depends on T , being minimum at T_0 (when the resonance condition is met). However, this way of probing exciton-cavity coupling is impractical when T_0 is low, because energy-level thermalization may then prevent light emission from the high-energy branch.

We have combined electric-field effects with the temperature dependence of the PL spectrum of a semiconductor microcavity to assess the degree of exciton-photon coupling at low temperature, when one of the them is not directly observable. The microcavity studied was formed by growing by molecular beam epitaxy dielectric mirrors on both sides of a semiconductor region that contained several isolated 80-Å quantum wells with exciton recombination wavelength λ . The mirrors consisted of twenty three pairs of layers, each pair formed by $\lambda/4$ regions of GaAlAs and AlAs. The optical cavity between the mirrors had a length of $3\lambda/2$, and housed one quantum-well pair at each of the antinode positions of the optical standing wave formed in the cavity.

Figure 3 shows representative photoluminescence spectra between 120K and 7K. As T decreases, the two peaks of the PL spectra shift to higher energy, but at different rates and with changing relative intensity. Below 40K only the low-energy structure is discernible. From the T dependence of the peaks, it is found that the one at the lower energy is mostly exciton-like while that at high energy is predominantly cavity-like. The fact that at the lowest temperatures the

latter disappears is consistent with a thermalized population of polaritons. However, how can we know whether there is exciton-cavity coupling?

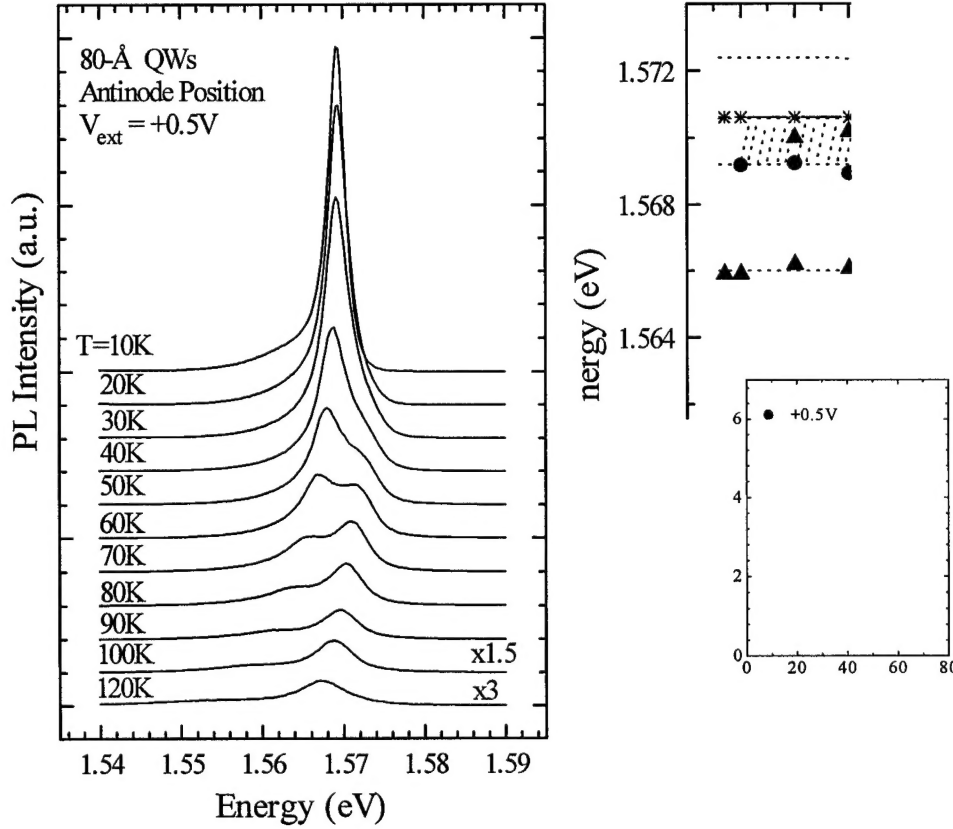


Fig. 3. (Left) Photoluminescence spectra at various temperatures for a microcavity in which 80-Å quantum wells are placed at antinode positions in the cavity.

Fig. 4. (Right) PL peak positions vs temperature for the spectra of Fig. 1(dots) and for spectra taken with the wells subject to an external electric field. (triangles). The asterisks correspond to peak positions for an 80-Å quantum well far away from resonance with the cavity.

We have shown that exciton-cavity coupling exists at low T by using the fact that an electric field can modify the well emission wavelength, and therefore the strength of the coupling. The dots in Fig. 4 indicate the peak energies of the spectra in Fig. 3, corresponding to the two polariton branches. The asterisks are the peak energies of an 80-Å well out of resonance with the optical cavity. The T dependence of the latter is similar to that of the polariton's low branch (presumably exciton-like) but not identical, the difference between them first increasing with decreasing T and then saturating at about 30K (dots in Fig. 4 inset). The triangles in Fig. 4 correspond to the microcavity's peak energies when an electric field of 60 kV/cm ($V_{\text{ext}} = -0.5$ V)

is applied between the cavity's dielectric mirrors. Although shifted, the low-energy peak runs parallel to that of the "bare" well (triangles in Fig. 4 inset). The position of the high-energy peak, on the other hand, deviates from the analogous peak at zero field.

These results are explained as follows. The electric field down-shifts the exciton's emission energy and effectively destroys the exciton-cavity coupling in spite of the existence of two PL peaks at moderate T . The high-energy peak for $F = 60$ kV/cm corresponds to the cavity mode in the absence of coupling, even at $F = 0$ (since the field does not directly affect the mode). Non interacting exciton and cavity modes would then approach each other as T decreases, and merge for $T \leq 40$ K. The coupling breaks that degeneracy in the microcavity at $F = 0$ and opens a 3 meV "gap," which is a measure of the interaction energy.

Personnel

The following people have been involved in this project:

- 1) Joong-Kon Son, Graduate Student
- 2) James Dickerson, Graduate Student
- 3) Fernando Camino, Graduate Student
- 4) Ralph Ruf, Engineer
- 5) Carlos Pecharromán, Postdoctoral Fellow
- 6) I.-Wei Tao, Postdoctoral Fellow
- 7) Emilio Mendez, Principal Investigator

In addition, Prof. Luis Viña, of Universidad Autónoma de Madrid, and his students Maria D. Martin and Guenther Aichmayr have been collaborators of the Stony Brook group in the time-resolved experiments.

Publications

1. *Signatures of Exciton-Cavity Coupling in Semiconductor Microcavities*, I. W. Tao, J. K. Son, C. Pecharromán, E. E. Mendez, and R. Ruf, *Physica E* **2**, 685 (1998)
2. *Tuning by an Electric Field of Spin Dependent Exciton-Exciton Interactions in Coupled Quantum Wells*, G. Aichmayr, M. Jetter, L. Viña, J. Dickerson, F. Camino, and E. E. Mendez, *Phys. Rev. Lett.* **83**, 2433 (1999)
3. *Spin Polarization Dynamics in a Semiconductor Microcavity*, M. D. Martin, L. Viña, R. Ruf, and E. E. Mendez, *Phys. Stat. Sol. (a)* **178**, 539 (1999)
4. *Spin-dependent Exciton-Exciton Interaction in Quantum Wells under an Electric Field*, G. Aichmayr, M. Jetter, L. Viña, J. Dickerson, F. Camino, and E. E. Mendez, *Phys. Stat. Sol. (b)* **215**, 223 (1999)

5. *Optical Gain of Type II based Quantum Cascade Lasers*, J. L. Jimenez and E. E. Mendez, Solid State Commun. **110**, 537 (1999)

Communications

1. *Temperature Dependence of Luminescence from Semiconductor Microcavities*, J. K. Son, I. W. Tao, C. Pecharroman, E. E. Mendez, and R. Ruf, Annual Meeting of American Physical Society, Los Angeles, March 16-20, 1998.
2. *Optical Gain in Type I and Type II Quantum Cascade Lasers*, E. E. Mendez and J. L. Jimenez, Annual Meeting of American Physical Society, Los Angeles, March 16-20, 1998.
3. *Dynamics of Polaritons and Stimulated Emission in GaAs Microcavities*, M. D. Martín, G. Aichmayr, L. Viña, J. K. Son, and E. E. Mendez, 24th Int. Conf. on the Physics of Semiconductors, Jerusalem, Israel, August 3-7, 1998.
4. *Quantum Wells and Superlattices for Optoelectronic Applications*, Opening Lecture, Scientific Inauguration of Materials Research Institute of Seville, January 14, 1998.

Interaction with Army Research Laboratory Scientists

We have maintained regular contacts with Drs. Richard Leavitt and John Bruno of the Army Research Laboratory. With Dr. Leavitt we discussed our project on field-induced band reversal; he grew in his MBE facility the InGaAs/InAlAs heterostructures used in the experimental part of the project. With Dr. Bruno we discussed our theoretical work on type II quantum cascade lasers. Dr. Bruno has an active interest in the subject, which has direct relevance to the Army Research Laboratory mission on communications and detection.

Honors

On October 23, 1998, in a solemn ceremony, the Principal Investigator received the Principe de Asturias Prize for Science and Technology, the most prestigious award among Spanish-speaking countries, for his pioneering work on semiconductor heterostructures, including the effects of an electric field on the optical properties of quantum wells and superlattices. This work has been partially supported by the ARO over the years.